Evaluation of the Population Distribution of Dietary Contaminant Exposure in an Arctic Population Using Monte Carlo Statistics

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Organochlorines and heavy metals have bioaccumulated in Arctic wildlife, which is an important food source for the Inuit. In this study, we have developed a statistical model to describe the population distribution of contaminant exposure and the usual intake of the high-end contaminant consumers. Monte Carlo methods are used to account for variations due to seasonal dietary pattern and contaminant concentrations. Distribution of the dietary intake of the contaminants of most concern—mercury, polychlorinated biphenyls (PCBs), chlordane, and toxaphenes—are described. Over 50% of the residents had dietary exposure levels exceeding the tolerable daily intake or provisional tolerable daily intake for Hg, toxaphene, and chlordane (83, 91, and 71% for men and 73, 85, and 56% for women, respectively). The high-end consumers (i.e. the 95th centile) have intake levels 6 times higher than the provisional tolerable weekly intake of Hg, and over 20 times the tolerable daily intake of chlordane and toxaphene. Assessment of health risks of the relative high contaminant exposure in this community must also consider the nutritional, economical, cultural, and social importance of these traditional foods. A comprehensive risk management scheme has yet to be developed. *Key words*: chlordane, exposure assessment, mercury, polychlorinated biphenyls, toxaphene, traditional diet. *Environ Health Perspect* 105:316–321 (1997)

Contaminants such as organochlorines and heavy metals are found in the Arctic environment as a result of long-range atmospheric and oceanic transport and local mining activities (*1*). Potential health effects on indigenous peoples are a concern because humans are at the top of the food chain and some of these pollutants are known to bioaccumulate (*2,3*). Results of preliminary dietary exposure assessment and maternal cord blood monitoring studies showed that some groups of indigenous people are exposed to high levels of contaminants such as toxaphene, polychlorinated biphenyls (PCBs), chlordane, mercury, and cadmium from the consumption of traditional food (*4–6*). We have previously reported the mean contaminant intake levels in an Arctic Inuit community of Qikiqtarjuaq on Baffin Island, Canada (*6–8*). The main purpose of these studies was to identify the contaminants of concern and their sources in the diet. It is, however, more important for risk assessors and risk managers that the distribution of exposure levels is characterized in detail [intake] and the usual intake levels of the high-end consumers are described. The usual intake is defined by Beaton (*9*) as "the average [daily] intake persisting over weeks or months, not days." The major issues of the number of overexposed individuals in the population and the intake of the high-exposure individuals still remain unanswered.

Conducting dietary exposure assessment in Arctic communities is a challenge because of the variation of intake due to seasonal availability of the food, family harvest variability, and large intrinsic variation of contaminants in the food. The latter factor is compounded by the limitation of sample size due to logistic reasons. Based on similar reasons, the use of statistical models for exposure/risk assessment has been gaining popularity in the last few years (*10–12*). Statistical methods for estimating usual exposure levels have been developed and evaluated by various researchers (*13–15*). However, there are no methods available that are entirely appropriate for the conditions of our data set, a data set which is typical of those collected from Arctic communities (i.e., marked seasonal variation, no more than one observation day per subject per season, and many subjects with missing data in many seasons). Therefore, an alternative, novel method was developed.

In this study, we present the development of a methodology for estimating the distribution of usual daily contaminant intake. This method addresses the variation of intakes due to seasonal dietary pattern and contaminant concentrations in traditional food to arrive at an estimate of usual daily contaminant intake. Using this methodology, we reanalyzed the Qikiqtarjuaq data and describe the population distribution of the intakes of mercury, PCBs, toxaphene, and chlordane.

To as full an extent as possible, the modeling, analyses, and presentation in this paper follow the fourteen principles "of good practice for the use of Monte Carlo Techniques" outlined by Burmaster and Anderson (*16*).

Materials and Methods

Contaminant exposures were estimated using three different methods, which we will refer to as 1-day, Point, and Monte Carlo estimates of contaminant intake. The dietary and contaminant data set collected between 1985 and 1988 in Qikiqtarjuaq, on Baffin Island, Canada, was used for the analyses. The methods used for the dietary study have been reported and are briefly described below:

Collection of dietary data. Twenty-four hour dietary recalls were conducted in Qikiqtarjuaq (population in 1987 = 586). Ninety adult females and 89 adult males were interviewed at least once, and at most six times, over a 1-year period. The number of days of data in consecutive 2-month periods starting July 1987 and ending June 1988 are 59, 51, 32, 51, 50 and 58 (total = 301) for males, and 71, 64, 61, 69, 69, and 67 (total = 401) for females . The patterns of traditional food consumption in the community have been previously described (*7*).

Measurement of contaminant levels in traditional foods. Samples of 90 traditional foods were collected in the forms normally consumed (e.g., raw, aged, boiled) by trained Inuit assistants. The samples were stored at -20°C, brought to our lab in Montreal, and analyzed using standard methods, as described previously (*7,8*). A data set containing mercury, chlordane, toxaphene, and PCB concentrations in 100 food items was established (*7,8*).

One-day estimates. The 1-day estimates of the intake of contaminant X (C_x) on each observed person-day were calculated as: Total $C_x = \sum(\text{food}_j \times c_{xj})$, where the intake of food$_j$ (in grams per day) is known from the dietary recall and the level of c_x in food$_j$ (in micrograms per gram) is the contaminant concentration measured. This is the most straightforward method and will provide an estimate of the average intake of

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We thank the Hamlet Council members, community members, and the many interviewers who participated. Funding for the initial community work was provided through Health Canada and the National Health Research and Development Program. This project was supported by the Arctic Environmental Strategy and the Department of Indian Affairs and Northern Development.

Received 27 August 1996; accepted 13 November 1996.

a population if enough day-person records are collected. This is the method used in our previous studies (*6–8*).

Point estimates. To estimate the usual annual intake of a person, an ideal analysis will use 2 or more days of dietary data from the person in each season, thus accounting for daily and seasonal variance in contaminant intake. A potential solution would be to take the individuals' average contaminant intakes over the six seasons. However, in the data set, only 9 males and 29 females had data available from all six seasons; thus an alternative strategy was employed.

Contaminant intake varied by season (e.g., mercury intake was highest in September 1987 and lowest in January 1988); however, no systematic correlation was observed between contaminant intake of individuals in different seasons. Multiple (matrix) correlation analyses were used to study the relationships between the contaminant intake in each of the six seasons. There were 120 correlations in total [contaminants (four) × sexes (two) × number of interseason comparisons (*15*)], but only 8 had a correlation coefficient with associated p-value <0.05 (6 would be expected to have a p-value <0.05 from chance alone). Therefore, while it was important to weigh each season equally in the estimation of usual intake, each day of intake in each season could be used as an independent data point, not linked to intake by that individual in other seasons.

The intake of C_x was calculated as above for the 1-day estimates: Total $C_x = \Sigma(\text{food}_j \times c_{xj})$. Then usual daily intakes of C_x over the entire year were estimated by taking the average of six C_x— one randomly selected C_x per season (randomly selected without replacement from the male or female set). This process was repeated until one season was exhausted [i.e., May 1988 with 32 (male) and 61 (female) days of data]. Therefore, 32 (male) and 61 (female) usual intakes could be calculated and the distribution of these intakes estimated. Because of the random differences that would be generated by selecting different random combinations from each season, this process was repeated until the distribution of distributions was stable [i.e., 50 repetitions; relative standard deviations (RSD) were about 10%].

Monte Carlo estimates. The methodology of the Point and 1-day estimates assumes that the contaminant concentrations (c_{xj}) are constant. In fact, contaminant concentrations in wildlife and fish parts vary between individuals within a species. Because the small sample size (usually one or two measures of c_x per food), we cannot accurately describe the distributions of c_x within each food. However, we have recently reviewed contaminant levels in the traditional food system across the Canadian Arctic and showed that between animals within a species, they are usually approximately lognormally distributed and with relative standard deviations of about 100% (*17*). Further support is garnered from additional analyses (Derek Muir, personal communication; Yukon Contaminants Committee, personal communication) of various metals and organochlorines in Arctic wildlife. Using the distribution-fitting software of Crystal Ball (Version 4.0, Decisioneering, Inc., Boulder, CO), we observed that lognormal distributions usually adequately describe the contaminant levels, with the Kolmogorov-Smirnov statistic for goodness-of-fit tests often less than 0.07 and usually less than 0.15. Although other distributions (e.g., normal, gamma) are better fitting in occasional situations, there is no systematic method for determining *a priori* in which situations distributions other than lognormal are more appropriate. We decided that using a lognormal distribution for all contaminants in all foods is an appropriate strategy until more refined data prove otherwise.

Therefore, we assume our measured level of c_{xj} is the mean, the RSD is 100%, and the distribution is lognormal. This distribution model was employed to generate an estimate of the distribution of intakes of C_x. The method used was similar to the Point method with the following differences. For each day of dietary data, C_x intakes were calculated 100 times as $C_x = \Sigma(\text{food}_j \times c_{xj})$, where the intake of food_j is known from the dietary recall and the level of C_x in food_j (in micrograms per gram) is randomly selected (with replacement) from the lognormal distribution (mean = c_x; RSD = 100%). One hundred repetitions were the maximum required to achieve consistent and stable distributions of contaminant intakes; therefore, there were between 3,200 and 5,900 days of data for adult males and between 6,100 and 7,400 days of data for adult females for each season. As described for the Point estimate, the usual intakes of C_x per day were estimated by taking the average of six C_x, one randomly selected C_x per season. This process was repeated until one season was exhausted [i.e., May 1988 with 3,200 (male) and 6,100 (female) days of data].

Exposure levels of mercury, chlordane, PCBs, and toxaphene from the consumption of traditional food were estimated using the three methods described. The exposure levels are compared to the guidelines established by Health Canada (V. Jerome, personal communication). For ease of comparison, the exposure levels were expressed as kilograms of body weight per day or week. The daily intakes were calculated and then divided by body weight (mean weight for the appropriate sex-age group from a previous survey of Canadian Inuit) (*18*).

Analyses were performed using SAS (Version 6.11, SAS Institute Inc., Cary, NC).

Results

Levels of exposure of Hg, PCBs, chlordane, and toxaphene are presented in Tables 1

Table 1. Summary of distribution of intakes of mercury, polychlorinated biphenyls (PCBs), chlordane, and toxaphene in Qikiqtarjuaq males

Contaminant	Mean ± SD	RSD (%)	50th percentile	95th percentile
Mercury (μg/kg/week)[a]				
1-day	13 ± 18	138	7	50
Point	13 ± 7	54	12	27
Monte Carlo	13 ± 10	77	11	33
PCBs (μg/kg/day)[b]				
1-day	0.85 ± 1.82	200	0.3	3.6
Point	0.85 ± 0.74	78	0.6	2.5
Monte Carlo	0.84 ± 0.94	113	0.6	2.4
Chlordane (μg/kg/day)[c]				
1-day	0.44 ± 1.2	273	0.06	2.1
Point	0.44 ± 0.42	95	0.30	1.5
Monte Carlo	0.44 ± 0.61	139	0.25	1.5
Toxaphene (μg/kg/day)[d]				
1-day	1.2 ± 3.6	300	0.05	8.1
Point	1.2 ± 1.1	92	0.65	3.9
Monte Carlo	1.1 ± 1.8	164	0.54	4.0

Abbreviations: SD, standard deviation; RSD, relative SD; TDI, tolerable daily intake.
The sample sizes for the 1-day, Point, and Monte Carlo methods are 301, 32, and 3,200, respectively.
[a]Provisional tolerable weekly intake = 5 μg/kg/week.
[b]Provisional tolerable daily intake = 1 μg/kg/day.
[c]TDI = 0.05 μg/kg/day.
[d]TDI = 0.2 μg/kg/day.

(males) and 2 (females). There are no differences in the mean levels of any of the four contaminants using the 1-day, Point, and Monte Carlo methods; however, the relative standard deviation was highest for the 1-day method, followed by the Monte Carlo method and the Point method. Results obtained from all three methods showed that males had higher levels of contaminant exposure than females. The average intake levels of mercury, chlordane, and toxaphene exceeded the guideline levels; provisional tolerable weekly intake (PTWI) for total mercury (0.05 μg/kg/week) established by the World Health Organization (WHO) (*19*), provisional tolerable daily intake (PTDI) for PCB (1 μg/kg/day) established by the Toxicological Evaluation Division of Health Canada (V. Jerome, personal communication), and tolerable daily intake (TDI) for chlordane (0.05 μg/kg/day) and toxaphene (0.2 g/kg/day) also established by the Toxicological Evaluation Division of Health Canada (personal communication).

Table 2. Summary of distribution of intakes of mercury, polychlorinated biphenyls (PCBs), chlordane, and toxaphene in Qikiqtarjuaq females

Contaminant	Mean ± SD	RSD (%)	50th percentile	95th percentile
Mercury (μg/kg/week)[a]				
1-day	11 ± 18	164	5	46
Point	11 ± 8	73	10	25
Monte Carlo	11 ± 10	91	8	31
PCBs (μg/kg/day)[b]				
1-day	0.62 ± 1.6	258	0.17	2.4
Point	0.62 ± 0.61	98	0.47	1.6
Monte Carlo	0.62 ± 0.82	132	0.40	1.8
Chlordane (μg/kg/day)[c]				
1-day	0.32 ± 1	313	0.04	1.4
Point	0.32 ± 0.34	106	0.21	0.91
Monte Carlo	0.32 ± 0.57	178	0.18	1.0
Toxaphene (μg/kg/day)[d]				
1-day	0.67 ± 2.2	328	0.04	4.2
Point	0.66 ± 0.86	130	0.34	2.1
Monte Carlo	0.65 ± 0.98	151	0.26	2.5

Abbreviations: SD, standard deviation; RSD, relative SD; TDI, tolerable daily intake.
The sample sizes for the 1-day, Point and Monte Carlo methods are 401, 61, and 6,100, respectively.
[a]Provisional tolerable weekly intake = 5 μg/kg/week.
[b]Provisional tolerable daily intake = 1 μg/kg/day.
[c]TDI = 0.05 μg/kg/day.
[d]TDI = 0.2 μg/kg/day.

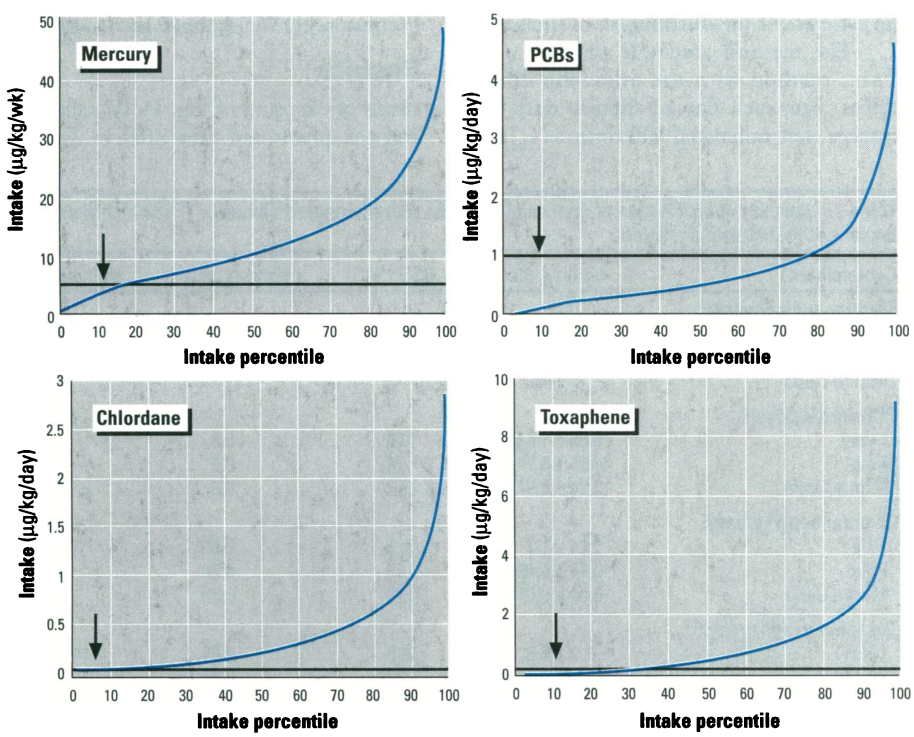

Figure 1. Estimation of population distribution of intake of mercury, polychlorinated biphenyls (PCBs), chlordane, and toxaphene for males using Bootstrap statistics. Arrows indicate provisional tolerable weekly intake for mercury = 5 μg/kg/week; provisional tolerable daily intake for PCBs = 1 μg/kg/day; and tolerable daily intakes for chlordane (0.05 μg/kg/day) and toxaphene (0.2 μg/kg/day).

The intake levels of the 50th percentile and 95th percentile are also shown in Tables 1 and 2. The 1-day method estimations always give the lowest values for the 50th percentile and the highest for the 95th percentile. Estimates obtained by the Point and Monte Carlo methods were similar. The median intake level obtained by the Point and Monte Carlo methods exceeded the guideline levels for Hg, chlordane, and toxaphene for both males and females.

Population distributions of the contaminants using the Monte Carlo method are shown in Figures 1 and 2. A sufficient numbers of iterations (100) were run so that a stable distribution was obtained (i.e., subsequent runs of the simulation will yield values ± 1% of those presented here). The guideline levels were also included in the figures. It is apparent that a high proportion of the population in the community had intake of Hg, chlordane, and toxaphene exceeding the guideline levels (Fig. 1, 2). The percentages of the community exceeding the guideline levels for all contaminants obtained by the three different methods are summarized in Table 3. Estimates obtained by the 1-day method were consistently the lowest, followed by those obtained by the Monte Carlo method and then those by the Point method. About 80% of the population had intakes higher than the guideline level of mercury, 20% had intakes higher than the guideline for PCBs, 90% had intakes higher than the guideline for chlordane, and 70% had intakes higher than the guideline for toxaphene.

It is also important to show the magnitude of overexposure among the high intake groups. We calculate hazardous indices by dividing the intake levels of the 95th percentile (from the Monte Carlo method) by the guideline levels; the results are summarized in Table 4. The high-end consumer had consumption levels at about 6 times the Hg guideline, about 2 times the PCB guideline, 30 times the chlordane guidelines and 20 times the toxaphene guideline.

Discussion

The mean levels of exposure estimated by the three different methods are similar and comparable to those described in previous reports (7,8). If only the mean exposure is required, then the 1-day method is appropriate because it is the simplest and the quickest method; however, the mean exposure is often of minimal value (although it is most often reported) because it does not adequately communicate the intake distribution, hence, the risk to the high-end consumer can not be evaluated.

Estimates obtained by the 1-day method showed highest variability because this method includes the day-to-day variations of the diet of the individual. For example, about 25% of the dietary records showed no intake of traditional foods and thus no measured intake of contaminants (lowering the 50th centiles). However, when traditional food was consumed, it often led to high single-day intakes of contaminants, showing higher 95th intake centiles. Both the Point and Monte Carlo methods estimate the usual intakes and reflect the reality that there are few, if any, people who have no traditional or a great deal of traditional food on a regular basis. Therefore, the distribution range is more narrow, i.e., the 50th centile is higher and the 95th is lower than the 1-day estimates.

From analysis of nutrient intake (20–23), it is known that the population distribution of a single day of data will be more widely distributed than the usual intakes. Except in cases of acute exposure, it is the usual intakes with which nutritionists and toxicologists are concerned; therefore, results generated from this method probably will not accurately represent the true usual distribution of contaminant intake. Nevertheless, it is included for comparison.

Estimation using the Monte Carlo method also showed lower 50th percentile levels and higher 95th percentile levels when compared to those obtained by the Point method; however, the differences are not as great as the 1-day method. The increase in variability is due to the incorporation of the intrinsic variations of the contaminant levels in the traditional foods by the Monte Carlo method; thus, the Monte Carlo method should be a better estimate of the true distribution. The error (uncertainty) in our estimates of the c_{xj} distribution is unknown. The method makes the assumption that the dietary data adequately reflect the variation in the intake, but the distribution of c_{xj} in the foods has to be estimated by modeling. Three assumptions are made in the model: 1) our measured concentrations are the means; 2) the RSD is 100%; and 3) the distribution is lognormal.

The Point estimates will of course have the same error if the first assumption is incorrect. Moreover, Beaton (24) has shown that for dietary nutrient intake, if the errors in the estimates of contaminant concentration in each food are random and many foods serve as a source of the nutrient (analogous to contaminant), then the estimates of total contaminant intake will not be too far off the true value (i.e., the underestimation of contaminant intake from one food will, to a large extent, be balanced with the overestimation of contaminant intake from another food). Effects on the population distribution by varying the RSD were investigated. An example showing the population distribution of chlordane is used to illustrate the effect of varying RSD from 50% to 150% (Fig. 3). The resulting variation of the 95th percentile estimates are less than 10%. Finally, lognormal distributions were observed for all four contaminants in over 50 species of wildlife and fish (17). Therefore, the use of Monte Carlo methods seems to be appropriate in the assessment of

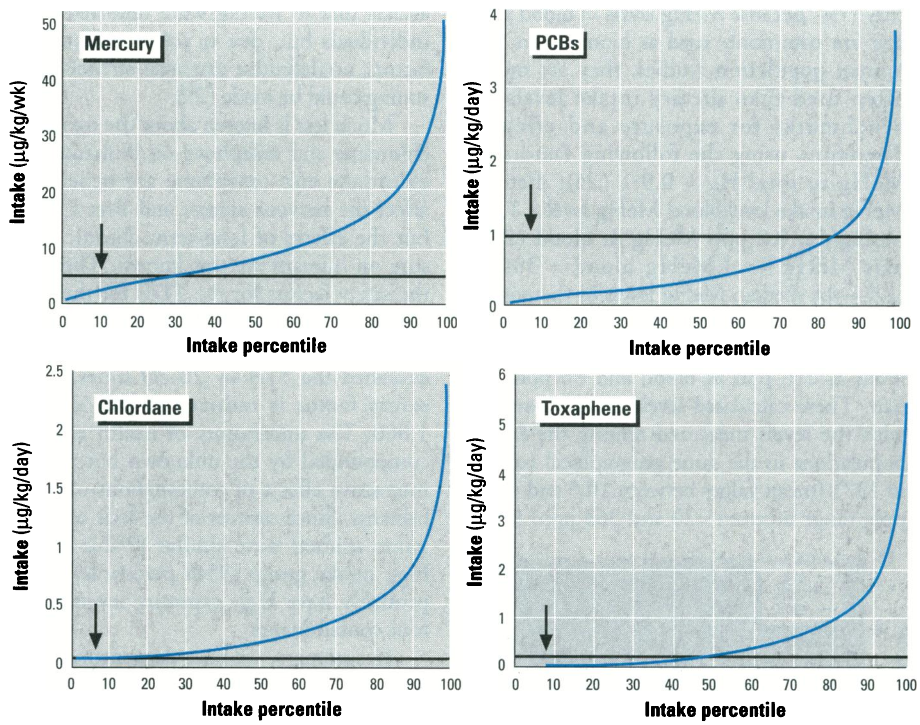

Figure 2. Estimation of population distribution of intake of mercury, polychlorinated biphenyls (PCBs), chlordane, and toxaphene for females using Monte Carlo statistics. Arrows indicate provisional tolerable weekly intake for mercury = 5 μg/kg/week; provisional tolerable daily intake for polychlorinated biphenyls (PCBs) = 1μg/kg/day; and tolerable daily intakes for chlordane (0.05 μg/kg/day) and toxaphene (0.2 μg/kg/day).

Table 3. Percentage of community with intakes of mercury, PCBs, chlordane and toxaphene greater than the guideline level

	Males			Females		
Contaminant	1-day	Point	Monte Carlo	1-day	Point	Monte Carlo
Mercury	67	90	83	49	80	73
PCBs	21	27	26	15	16	15
Chlordane	52	93	91	47	89	85
Toxaphene	32	75	71	26	60	56

Table 4. Hazardous index[a] showing the magnitude of overexposure to contaminants in traditional food

Contaminant	Males			Females		
(safety factor in PTDI)	1-day	Point	Monte Carlo	1-day	Point	Monte Carlo
Mercury (1)	10	5	7	9	5	6
PCBs (100)	4	3	2	2	2	2
Chlordane (1000)	43	30	30	27	18	21
Toxaphene (1000)	40	20	20	21	11	12

Abbreviations: PTDI, provisional tolerable daily intake; PCBs, polychlorinated biphenyls.
[a]Hazardous index is defined as (95th intake percentile)/tolerable daily intake.

contaminant exposure in the Arctic where both the dietary and contaminant data are limited.

Chronic exposure of methylmercury (MeHg) has been a major concern among fish-eating populations including indigenous communities in Canada. In the Inuit community that we studied, marine mammal tissues are the main sources of all four contaminants studied (Table 5). The PTWI for total Hg has been set at 5 µg/kg/week of which no more than 3 µg/kg/week may be methylmercury (*19*). Because MeHg levels in blood and hair are commonly used as biomonitors for human population studies, they are more often used than dietary intake levels as benchmarks for exposure and effects. Therefore, using the following factors—MeHg to total Hg = 0.9:1 (*25*); dietary MeHg intake level:blood MeHg level = 1 µg MeHg/day:0.8 ppb MeHg in blood (*26*); hair MeHg level:MeHg blood = 300:1 (*27*)—the median MeHg levels in the population can be calculated as 73 ppb in blood and 22 ppm in hair and the 95th percentile levels as 216 ppb in blood and 66 ppm in hair. These calculated levels are comparable with the levels measured among the Inuit populations in the same geographical region in 1971 (mean values between 10.5 and 49.5 ppb; range between <20 and 363 ppb) (*28*).

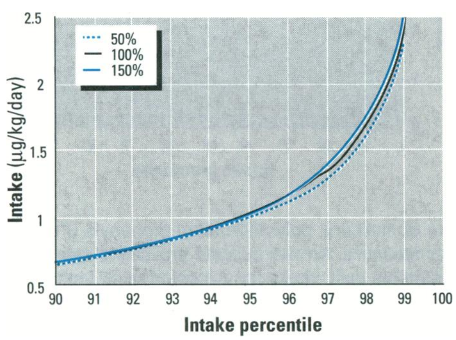

Figure 3. Estimation of population distribution of intake of toxaphene for female assuming that the relative standard deviations of the distribution of toxaphene concentrations in traditional food are 50, 100, and 150%.

Based on the Iraqi data, the WHO suggests that there is a 5% risk of neonatal neurological disorders associated with a peak MeHg level of 10–20 ppm in maternal hair (*19*). Moreover, 5% of the adult population may be at risk of developing early signs of Hg toxicity (paraesthesia) at a blood Hg level of 200 ppb. Therefore, both dietary and biomonitoring studies indicate the potential problem of high Hg exposure in the Inuit population in Northern Canada. Among indigenous peoples in Canada, possible neurological effects due to MeHg were reported in 11 individuals but, due to other confounding factors, no definitive diagnosis of MeHg poisoning could be made (*29*).

Much less is known about the toxicity of chlordane and toxaphene on humans. Both chlordane and toxaphene are believed to affect the nervous system and liver (*30,31*), but the effects of long-term chronic exposure on humans are not known. Therefore, the safety factors for the TDIs for these two classes of compounds are set at 1,000. Our results show that the 95th percentile intake exceeded the TDI by 20–30 times, or the safety factor is reduced to 30–50 from 1,000. The uncertainty of health effects is compounded by the unknown but possible interactive effects of combinations of contaminants. Since sources of the four contaminants studied were similar (Table 5), the high intake group (95th percentile) would probably have high exposure levels for all four contaminants.

In summary, we have demonstrated that Monte Carlo statistics are useful tools to estimate the distribution of contaminant exposure in an Arctic community. Results of the population distribution present a much better picture on the magnitude of overexposure of Hg, chlordane, and toxaphene in comparison to the guideline levels. A quantitative risk assessment on the public health impact is required. Moreover, given the known nutritional, economical, cultural, and social importance of traditional foods in the Arctic Inuit community (*32*), a comprehensive risk management scheme involving the local people is also needed. Results of this study were communicated to the community in September 1996. The Inuit populations are aware of the situation and are asking for more information in order to make better informed choices.

Table 5. The top three traditional food items contributing to contaminant exposure by Baffin Inuit women >20 years old in Qikiqtarjuaq

	Top three food items		
Contaminant	1st	2nd	3rd
Mercury	Ringed seal meat (41)	Narwhal mattak (22)	Ringed seal liver (12)
PCBs	Walrus blubber (29)	Narwhal mattak (16)	Ringed seal blubber (13)
Chlordane	Walrus blubber (37)	Ringed seal blubber (16)	Narwhal blubber (16)
Toxaphene	Narwhal blubber (45)	Walrus blubber (23)	Narwhal mattak (16)

PCBs, polychlorinated biphenyls.
Values in parentheses are contributions to total exposure (%). Adapted from Kinloch et al. (*6*) and Chan et al. (*7*).

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